SYNTHETIC AND STRUCTURAL STUDIES WITH SOME SUBSTITUTED ORGANIC SULFONAMIDES AND ALLIED COMPOUNDS

A SYNOPSIS

OF THE THESIS TO BE SUBMITTED

TO

THE FACULTY OF SCIENCE AND TECHNOLOGY OF

MANGALORE UNIVERSITY

FOR AWARD OF THE DEGREE OF

DOCTOR OF PHILOSOPHY

IN

CHEMISTRY

BY

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August 2011

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Title of the Thesis **: SYNTHETIC AND STRUCTURAL STUDIES WITH SOME SUBSTITUTED ORGANIC SULFONAMIDES AND ALLIED COMPOUNDS**

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The thesis describes the results of synthetic, spectral and structural studies on a large number of substituted maleamic acids, maleic diamides, N-(arylsulfonyl)-substitutedacetamides and N-haloarylsulfonamides. The objective of the research is to study the effect of substituents on the geometrical parameters and to correlate the spectroscopic parameters with the bond parameters of these compounds. It also describes the effect of substituents on the hydrogen bond patterns observed in this class of compounds.

Spectroscopy and its applications form a significant part of modern chemistry and physics[1]. Spectroscopic technique is one of the most powerful tools available for the study of atomic and molecular structures and is used in the investigation of a wide range of samples. Infrared spectroscopy provides useful information about the structure of a molecule quickly and convincingly without tiresome evaluation methods. The direct applications of NMR studies are to provide evidences relating to the nature of the chemical bonding in molecules or crystals. Advances in the NMR techniques have led to wide spread applications in various branches of science, which include chemical, medical, solid state and biophysical sciences. Knowledge of structure of materials is essential for a proper understanding of their physical and chemical properties. Thus crystal structure studies have been extensively used to investigate the structural aspects of a variety of compounds[2, 3].

Amides are one of the important classes of organic compounds in which the hydroxy groups of carboxylic or sulfonic acids are replaced by amino groups. The structure and properties of the amide group have been the subject of much interest. The versatility of the amide group in forming partial bonds with itself and many other functional groups is partly responsible for the structural subtleties of the biologically important amino acids, peptides and proteins etc., which are fundamental to all life. These can be understood in terms of delocalisation of the nitrogen lone pair electron into the π - system of the carbonyl group. The latter produce partial double bond character in the -C(O)-N bond and generate a 1,3 - dipole, with nitrogen bearing a partial positive charge and oxygen, the partial negative charge. Consequences of partial double bond character are the planar nature of the amide group and the existence of configurational isomers. The donor-acceptor properties of the amide moiety manifest in acid-base and complexing interactions and a tendency to self associate are a consequence of its dipolar structure.

Sulfonamides and their analogues show distinct physical, chemical and biological properties. Many aryl sulfonamides and their N-halo compounds also exhibit pharmacological, fungicidal and herbicidal activities due to their oxidising action in aqueous, partial aqueous and non-aqueous media[4,5]. The chemistry of these compounds is unique in that they are

sources of halonium cations, hypohalite species and N-anions which act both as bases and nucleophiles. These potential reagents react with a wide range of functional groups in aqueous and partial aqueous media in the presence of acids or alkalies effecting an array of molecular transformations. N-Halo compounds are also of interest in synthetic, mechanistic, analytical and biological chemistry. Their oxidising power depends on the ease with which the halogen is released as positive ion in the reaction system, which in turn depends on the electron environment around the N-X bond. Thus a number of N-(halo)-aryl/alkyl-sulfonamides of different oxidising strengths have been introduced for varied applications. Some of them have been employed as oxidants for studying the kinetics of reactions of substrates of industrial and biological interest[5].

As a part of efforts of correlating spectroscopic parameters with the chemical bond parameters, a number of substituted maleamic acids of the general formula i-$XC_6H_4NHCOCH=CHCOOH$ (i = 2, 3 or 4 and X = Cl, Br, F, CH_3, NO_2 or OCH_3), i,j-$X_2C_6H_3NHCOCH=CHCOOH$ (i,j = 2,3; 2,4; 2,5; 2,6; 3,4 or 3,5 and X = Cl , CH_3 or NO_2), i,j,k-$X_3C_6H_2NHCOCH=CHCOOH$ (i,j,k = 2,3,4; 2,4,5 or 2,4,6 and X = Cl or CH_3), i-X,j-$YC_6H_3NHCOCH=CHCOOH$ (i or j = 2, 4 or 5 and X or Y = Cl, CH_3 or NO_2) and 2,6-Cl_2,4-$YC_6H_2NHCOCH=CHCOOH$ (Y = Br or NO_2) have been prepared, characterized and their infrared spectra in the solid state and ^1H & ^{13}C NMR spectra in solution measured and correlated. Further, maleic diamides of the general formula, i-$XC_6H_4NHCOCH=CHCOHNC_6H_4$i-X (i = 2, 3 or 4 and X = H, Cl or CH_3) have also been prepared, characterized and their infrared and ^1H & ^{13}C NMR spectra are measured and correlated. Several N-(aryl)-4-chloro/methylbenzenesulfonamides have also been prepared, characterized and their crystal structures have been determined and analyzed. A number of N-(arylsulfonyl)-substitutedacetamides of the general formula, 2-$YC_6H_4SO_2NHCOCH_{3-i}X_i$ (X, Y = CH_3 or Cl and i = 0, 1, 2, or 3); metal salts of N-haloarylsulfonamides and N, N-dihaloarylsulfonamides of the general formulae, 2-$YC_6H_4SO_2NXNa/K$ and 2-$YC_6H_4SO_2NX_2$ (Y = Cl or CH_3 and X = Cl or Br) have been prepared, characterized and their infrared and ^1H & ^{13}C NMR spectra are measured and correlated.

Single - Crystal X-ray structures of several of the above compounds and a number of N-arylsulfonamides of the general formula, 4-YC_6H_4-SO_2NH-(i-XC_6H_4/i,j-$X_2C_6H_3$) (X = H, Cl or CH_3; Y = Cl or CH_3; i = 2, 3 or 4 & i,j = 2,3; 2,4; 2,5; 2,6; 3,4 or 3,5) have been determined and analyzed to explore the effect of substituents on the geometrical parameters of these compounds.

Infrared spectral measurements were made on FT-IR spectrophotometers. The resolution was set to 4 cm^{-1} and the scanning range was from 400 to 4000 cm^{-1}. The spectra were measured in the solid state in the form of KBr pellets.

The NMR spectral measurements were carried out on a BRUKER Ac 300F, 400MHz FT-NMR spectrometer. Both ^{1}H and ^{13}C NMR spectra were recorded in $CDCl_3$/DMSO with tetramethylsilane (Me_4Si) as internal standard.

Small single crystals of the compounds used in X-ray diffraction studies were grown by slow evaporations of dilute solutions of the recrystallized samples of the compounds in ethanol or aqueous ethanol. The single-crystal X-ray studies were carried out mostly at room temperature. The collected intensity data were corrected for Lorentz polarisation and absorption. The positional parameters were determined by direct methods and least squares refinement (SHELXL-97)[2].

The details of structure solutions are as follows[2] :

Measurement device: CAD-4 Diffractometer (Enraf–Nonius, 1996)
Xcalibur Diffractometer with Sapphire CCD detector (Oxford Diffraction, 2009).

Data collection: *CAD-4-PC* (Nonius, 1996)
CrysAlis CCD (Oxford Diffraction, 2009);

Cell refinement: *CAD-4-PC* (Nonius, 1996)
CrysAlis RED (Oxford Diffraction, 2009);

Data reduction: REDU4 (Stoe & Cie, 1987)
CrysAlis RED (Oxford Diffraction, 2009);

Program(s) used to solve structure: *SHELXS97* (Sheldrick, 2008);

Program(s) used to refine structure: *SHELXL97* (Sheldrick, 2008);

Molecular graphics: *PLATON* (Spek, 2003); *ORTEP*-3 (Farrugia, 1997)
DIAMOND (Brandenburg, 2002);

Software(s) used: *SHELXL97* (Sheldrick, 2008); *PLATON* (Spek, 2009)
WinGX (Farrugia,1999).

The results of most of these investigations have been published and the remaining results are also at various stages of publication[6-12]. Results of these synthetic, spectral and structural studies are presented and discussed in the thesis. The thesis is broadly divided into TEN CHAPTERS with several sections in each chapter. The list of contents of the thesis are as follows:

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CHAPTER 10: CRYSTAL STRUCTURES OF N-ARYLSULFONAMIDES AND SODIUM SALTS OF N-CHLORO-ARYLSULFONAMIDES

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10.2 Crystal Structures of o-Substitutedbenzenesulfonamides

10.3 Crystal Structures of Sodium Salts of N-Chloro-o-substituted-benzenesulfonamides

Chapter 1 gives an introduction to the infrared and NMR spectroscopic techniques, crystal structure studies, chemistry of arylsulfonamides and sketches the objectives and scope of the present work.

Chapter 2 describes the synthetic, infrared, ^{1}H and ^{13}C NMR spectral studies on N-aryl maleamic acids along with the review of literature. The infrared spectra of 40 N-(aryl)maleamic acids have been measured in the solid state. The variations of IR absorptions with the substitution in the benzene ring are shown in terms of line diagrams. The ^{1}H and ^{13}C NMR spectra of the above compounds have also been studied. The chemical shifts of aromatic protons and carbons are calculated by four different methods and compared with the measured values. There is a good agreement between the measured and calculated values.

Chapter 3 describes the effect of substituents on the crystal structures of twelve N-(aryl)maleamic acids. The crystal structures of N-(2-methylphenyl)maleamic acid, N-(3-chlorophenyl)maleamic acid, N-(3-nitrophenyl)maleamic acid, N-(4-methoxyphenyl)-maleamic acid, N-(2,6-dimethylphenyl)maleamic acid, N-(3,4-dimethylphenyl)maleamic acid, N-(2,5-dichlorophenyl)maleamic acid, N-(3,4-dichlorophenyl)maleamic acid, N-(3,5-dichlorophenyl)-maleamic acid, N-(2,4,6-trimethylphenyl)maleamic acid, N-(2,4,5-trichlorophenyl)maleamic acid and N-(2,4,6-trichlorophenyl)maleamic acids have been determined. The data have been analyzed along with those of N-(phenyl)maleamic acid, N-(4-methylphenyl)maleamic acid, N-(4-chlorophenyl)maleamic acid and N-(4-nitrophenyl)-maleamic acid, reported in literature, to explore the effect of the nature, site and the quantum of substitutions on the structural parameters of these compounds.

Chapter 4 describes the synthetic, infrared, ^{1}H & ^{13}C NMR spectral and crystal structure studies on maleic diamides. The infrared and ^{1}H & ^{13}C NMR spectra of seven maleic diamides of the general formula i-$XC_6H_4NHCOCH{=}CHCOHNC_6H_4$i-X (i = 2, 3 or 4 and X = H, Cl or CH_3) have been measured and analysed. The crystal structures of N, N-Bis(phenyl)-maleamide and N, N-Bis(4-Chlorophenyl)-maleamide have also been described.

Chapter 5 describes the synthetic, infrared, ^{1}H and ^{13}C NMR spectral studies on 14 N-(orthosubstitutedphenylsulfonyl)-substitutedacetamides of the general formula, 2-$YC_6H_4SO_2NHCOCH_{3-i}X_i$ (X /Y= CH_3 or Cl and i = 0, 1, 2 or 3), along with a review of literature on the title compounds. The infrared spectra of N-(o-substitutedphenylsulfonyl)-substitutedacetamides have been measured in the solid state. The ^{1}H and ^{13}C NMR chemical shifts (δ, ppm) of C=O, C-1 and N-H groups in N-(2-chloro/2-methylphenylsulfonyl)-

substitutedacetamides have been compared with those of N-(phenylsulfonyl)-substitutedacetamides and N-(4-chloro/4-methyl/4-bromophenylsulfonyl)-substituted-acetamides.

Chapter 6 describes the crystal structures of eight N-(o-substitutedphenylsulfonyl)-acetamides / substitutedacetamides of the general formula, 2-$YC_6H_4SO_2NHCO(CH_{3-i}X_i)$ (X, Y = CH_3 or Cl and i = 0, 1, 2 or 3) to consider the effect of substituents on the structures of these compounds. Although, with the limited data, it is difficult to generalise the effect of substituents on the unit cell and the bond parameters, it is evident from the comparison of data that the nature and quantum of substitutions in the side chain significantly alter the structural parameters.

Chapter 7 discribes the crystal structures of 6 N-(dichlorophenyl)-4-methyl-benzenesulfonamides and analyses the data to know the effect of substituents on the structural parameters of these compounds. The crystal structures of N-(2,3-dichlorophenyl)-4-methylbenzenesulfonamide, N-(2,4-dichlorophenyl)-4-methylbenzenesulfonamide, N-(2,5-dichlorophenyl)-4-methylbenzenesulfonamide, N-(2,6-dichlorophenyl)-4-methylbenzene-sulfonamide, N-(3,4-dichlorophenyl)-4-methylbenzenesulfonamide and N-(3,5-dichloro-phenyl)-4-methylbenzenesulfonamide have been determined.

Chapter 8 discusses the effect of substituents on the crystal structures of N-(aryl)-4-chlorobenzenesulfonamides. The crystal structures of N-(phenyl)-4-chlorobenzenesulfonamide, N-(3-methylphenyl)-4-chlorobenzenesulfonamide, N-(2-chlorophenyl)-4-chlorobenzene-sulfonamide, N-(3-chlorophenyl)-4-chlorobenzenesulfonamide, N-(2,3-dimethylphenyl)-4-chlorobenzenesulfonamide, N-(2,4-dimethylphenyl)-4-chlorobenzenesulfonamide, N-(2,5-dimethylphenyl)-4-chlorobenzenesulfonamide, N-(2,6-dimethylphenyl)-4-chlorobenzene-sulfonamide, N-(3,5-dimethylphenyl)-4-chlorobenzenesulfonamide, N-(2,3-dichlorophenyl)-4-chlorobenzenesulfonamide and N-(3,5-dichlorophenyl)-4-chlorobenzenesulfonamide have been determined. The comparison and analysis of the structural data have indicated that the position and nature of substitution alter the unit cell and geometrical parameters.

Chapter 9 describes the synthetic, infrared and ^1H & ^{13}C NMR spectral studies on N-haloarylsulfonamides and N, N-dihaloarylsulfonamides along with the review of literature. The variations of $\bar{\nu}_{S=O}$, $\bar{\nu}_{N-Cl}$ & $\bar{\nu}_{N-Br}$ absorptions of 2-$YC_6H_4SO_2NaNX$ and 2-$YC_6H_4SO_2NX_2$ (Y = H, Cl or CH_3 and X = Cl or Br) are compared with those of 4-$YC_6H_4SO_2NaNX$ and 4-$YC_6H_4SO_2NX_2$ (Y = Cl, CH_3, F, Br or NO_2 and X = Cl or Br).

Chapter 10 discusses the crystal structures of N-aryl and metal salts of N-halo-aryl-sulfonamides. The crystal structures of N-2-chlorobenzenesulfonamide, N-2-methylbenzene-sulfonamide, and sodium salts of N-chloro-2-chlorobenzenesulfonamide and N-chloro-2-methyl-benzenesulfonamide have been determined. The data have been analyzed along with those of the parent and the corresponding para-substituted compounds.

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12. B. T. Gowda, S. Foro, K. Shakuntala and H. Fuess , *Acta Cryst.* **E67** (2011) o117.

13. B. T. Gowda, S. Foro and K. Shakuntala *Acta Cryst.* **E67** (2011) o1569, m870, m914, m926, m961, m962, m1015.

Attested by the Guide

(Mrs. K. Shakuntala)

Prof. B. Thimme Gowda Signature of the Candidate

LIST OF PUBLICATIONS

1. Structure Report on 2-Chlorobenzenesulfonamide
   B. T. Gowda, S. Foro, **K. Shakuntala** and H. Fuess, *ActaCryst*. **E65** (2009) o2144

2. Structure Report on o-Toluenesulfonamide: a redetermination
   B. T. Gowda, S. Foro, **K. Shakuntala** and H. Fuess, *ActaCryst*. **E65** (2009) o2258

3. Structure Report on N-(2, 6-Dimethylphenyl)maleamic acid
   B. T. Gowda, M. Tokarcik, J. Kozisek, **K. Shakuntala** and H.Fuess
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4. Structure Report on, N-(3,4-Dimethylphenyl)maleamic acid
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